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# Spinodal instability in asymmetric nuclear matter

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We demonstrate that the instabilities of asymmetric nuclear matter at sub-saturation densities do not present two types of instabilities as usually discussed but a unique one. The associated order parameter is everywhere dominated by the isoscalar density and so the transition is of liquid-gas type even in the so-called chemical instability region. However, the instability goes in the direction of a restoration of the isospin symmetry leading to a fractionation phenomenon.

## 1. Introduction

Below saturation density, the nuclear interaction is expected to lead to a liquid-gas phase transition [1]. Recently, a converging ensemble of experimental signals seems to have established the phase transition. One is the spinodal decomposition [2]. It has been argued that asymmetric nuclear matter do not only present a mechanical instability for which the density is the order parameter but also a broader chemical instability associated with fluctuations of the matter isospin content [4]. Indeed, it is usually argued that it exists a region in which the compressibility at constant asymmetry and constant isospin is negative (see figure (1)) leading to the interpretation that the system is mechanically unstable. Above a maximum instability the isotherms at constant asymmetry does not presents any back bending leading to the idea that the system is mechanically stable. However, looking at the equilibrium of the chemical potentials one can see that above this maximum asymmetry for mechanical instabilities the system may amplify fluctuations in the proton neutron concentration leading to a second instability region usually called chemical instabilities.

However, we have recently shown that this splitting of the spinodal region in to two types of instabilities a mechanical and a chemical one is not correct and that ANM present only one type of instabilities [3]. The associated order parameter is dominated by the isoscalar density and so the transition is of liquid-gas type. The instability goes in the direction of a restoration of the isospin symmetry leading to a fractionation phenomenon. Our conclusions are model independent since they can be related to the general form of the asymmetry energy. They are illustrated using density functional approaches derived from Skyrme and Gogny effective interactions.

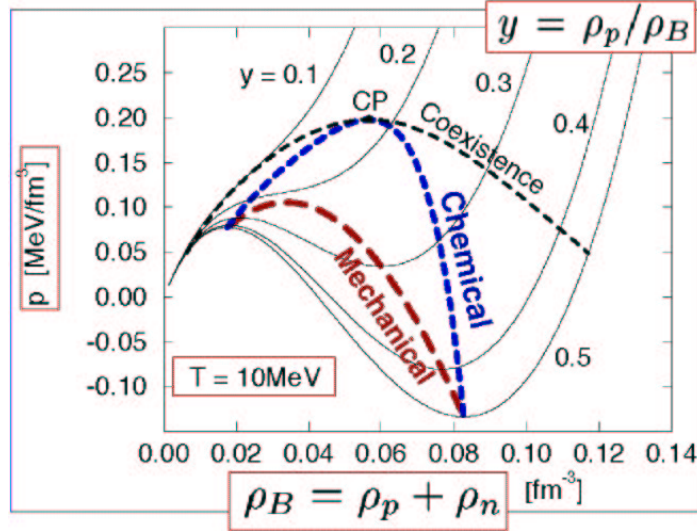


Figure 1. This figure represents the pressure as a function of the total density for a fixed temperature and different asymmetry. The region where the isotherms present a back bending are usually related to a mechanical instability. However, one can define a broader region of instability by allowing the isospin fluctuations. This region of proton and neutron concentration instability is traditionally discussed as a chemical instability. We will show that this classification is artificial since it does not correspond to a modification of the physical properties of the instabilities.

## 2. Stability analysis

Let us consider ANM characterized by a proton and a neutron densities  $\rho_i = \rho_p, \rho_n$ . These densities can be transformed in a set of 2 mutually commuting charges  $\rho_i = \rho_1, \rho_3$  where  $\rho_1$  is the density of baryons,  $\rho_1 = \rho_n + \rho_p$ , and  $\rho_3$  the asymmetry density  $\rho_3 = \rho_n - \rho_p$ . In infinite matter, the extensivity of the free energy implies that it can be reduced to a free energy density :  $F(T, V, N_i) = V\mathcal{F}(T, \rho_i)$ . The system is stable against separation into two phases if the free energy of a single phase is lower than the free energy in all two-phases configurations. This stability criterium implies that the free energy density is a convex function of the densities  $\rho_i$ . A local necessary condition is the positivity of the curvature matrix :

$$[\mathcal{F}_{ij}] = \left[ \frac{\partial^2 \mathcal{F}}{\partial \rho_i \partial \rho_j} \Big|_T \right] \equiv \left[ \frac{\partial \mu_i}{\partial \rho_j} \Big|_T \right] \quad (1)$$

where we have introduced the chemical potentials  $\mu_j \equiv \frac{\partial F}{\partial N_j} \Big|_{T, V, N_i} = \frac{\partial \mathcal{F}}{\partial \rho_j} \Big|_{T, \rho_{i \neq j}}$ .

We represent in Fig. 2 the energy surface as a function of  $\rho_n$  and  $\rho_p$ , deduced from SLy230a Skyrme interaction [13]. In the symmetric case ( $\rho_n = \rho_p$ ), one can see the negative curvature of the energy which defines the spinodal area, whereas in pure neutron matter ( $\rho_p = 0$ ), no negative curvature and so no spinodal instability are predicted. We can also notice that the isovector density dependence is almost parabolic illustrating the positivity of  $\mathcal{F}_{33}$ .

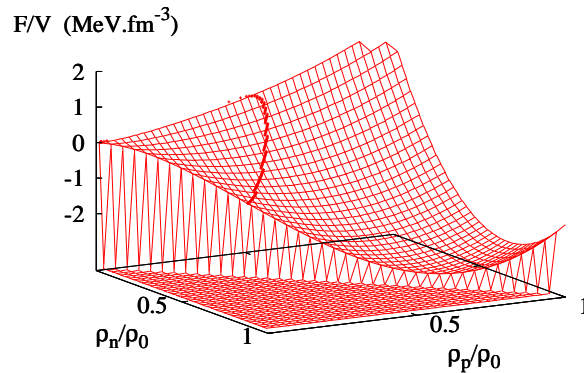


Figure 2. This figure represents the energy surface as a function of the densities  $\rho_n$  and  $\rho_p$  for the SLy230a interaction. The contour delimitate the spinodal area.

We show in the left part of Fig. 3 the spinodal contour in ANM for several forces. It exhibits important differences. In the case of SLy230a force (as well as SGII, D1P), the total density at which spinodal instability appears decreases when the asymmetry increases whereas for SIII (as well as D1, D1S) it increases up to large asymmetry and finally decreases. We observe that all forces which fulfill the global requirement that they reproduce symmetric nuclear matter (SNM) equation of state as well as the pure neutron matter calculations, leads to the same curvature of the spinodal region. We can

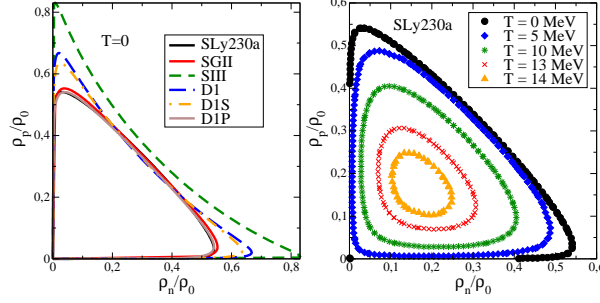


Figure 3. This two figures are a projection of the spinodal contour in the density plane : left, for Skyrme (SLy230a [13], SGII [14], SIII [15]) and Gogny models (D1 [12], D1S [17], D1P [18]) ; right, temperature dependence of the spinodal zone computed for the SLy230a case.

appreciate the reduction of the instability when we go away from SNM. However, large asymmetries are needed to induce a sizable effect. The temperature dependence of the spinodal contour can be appreciated in the right part of Fig. 3. As the temperature increases the spinodal region shrinks up to the critical temperature for which it is reduced to SNM critical point. However, up to a rather high temperature ( 5 MeV) the spinodal zone remains almost identical to the zero temperature one.

Almost all theoretical predictions has been made with simplified Skyrme interactions : in medium nucleonic masses are taken as the free masses and exchange terms are not explicitly treated. In our case, we have included the standard terms of the interactions that we refer to. Fig. 4 shows a comparison between one of those simplified interactions (used by V.Baran [7]) and our calculation. The critical temperature is reduced by 1 MeV and the behaviour with the asymmetry is slightly different. Anyway, this comparison shows that for qualitative predictions, this simple interaction is enough but quantitative predictions requires the standard interaction.

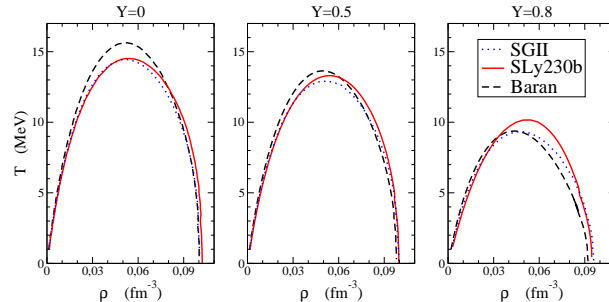


Figure 4. comparison between the results obtained by Skyrme simplified Baran interaction and the one obtained with SLy230b Skyrme interaction.

### 3. Analysis of the curvature matrix $[\mathcal{F}_{ij}]$

In the considered two-fluids system, the  $[\mathcal{F}_{ij}]$  is a  $2 \times 2$  symmetric matrix, so it has 2 real eigenvalues  $\lambda^\pm$  [7] :

$$\lambda^\pm = \frac{1}{2} \left( \text{Tr} [\mathcal{F}_{ij}] \pm \sqrt{\text{Tr} [\mathcal{F}_{ij}]^2 - 4 \text{Det} [\mathcal{F}_{ij}]} \right) \quad (2)$$

associated to eigenvectors  $\delta\rho^\pm$  defined by ( $i \neq j$ )

$$\frac{\delta\rho_j^\pm}{\delta\rho_i^\pm} = \frac{\mathcal{F}_{ij}}{\lambda^\pm - \mathcal{F}_{jj}} = \frac{\lambda^\pm - \mathcal{F}_{ii}}{\mathcal{F}_{ij}} \quad (3)$$

Eigenvectors associated with negative eigenvalue indicate the direction of the instability. It defines a local order parameter since it is the direction along which the phase separation occurs. The eigen values  $\lambda$  define sound velocities,  $c$ , by  $c^2 = \frac{1}{18m}\rho_1 \lambda$ . In the spinodal area, the eigen value  $\lambda$  is negative, so the sound velocity,  $c$ , is purely imaginary and the instability time  $\tau$  is given by  $\tau = d/|c|$  where  $d$  is a typical size of the density fluctuation.

The requirement that the local curvature is positive is equivalent to the requirement that both the trace ( $\text{Tr}[\mathcal{F}_{ij}] = \lambda^+ + \lambda^-$ ) and the determinant ( $\text{Det}[\mathcal{F}_{ij}] = \lambda^+ \lambda^-$ ) are positive

$$\text{Tr}[\mathcal{F}_{ij}] \geq 0, \text{ and } \text{Det}[\mathcal{F}_{ij}] \geq 0 \quad (4)$$

The use of the trace and the determinant which are two basis-independent characteristics of the curvature matrix clearly stresses the fact that the stability analysis should be independent of the arbitrary choice of the thermodynamical quantities used to label the state e.g.  $(\rho_p, \rho_n)$  or  $(\rho_1, \rho_3)$ .

### 4. Link with the usual definition of chemical or mechanical instabilities

A different discussion can be found in the literature [4,5] and we will now clarify the relation of this discussion with the eigen modes analysis. Indeed, from the thermodynamical relation

$$\rho_n \text{Det}[\mathcal{F}_{ij}] = \frac{\partial\mu_p}{\partial y}|_{T,P} \frac{\partial P}{\partial\rho_1}|_{T,y} \quad (5)$$

one can be tempted to relate separately  $\frac{\partial\mu_p}{\partial y}|_{T,P}$  and  $\frac{\partial P}{\partial\rho_1}|_{T,y}$  to the two eigenvalues  $\lambda^+$  and  $\lambda^-$ . The discussion of the sign of  $\lambda^+$  and  $\lambda^-$  is replaced by the discussion of the sign of  $\frac{\partial\mu_p}{\partial y}|_{T,P}$  (also called chemical instability) and  $\frac{\partial P}{\partial\rho_1}|_{T,y}$  (also called mechanical instability). This is correct in SNM and the mechanical instability is associated with density fluctuations whereas the chemical instability is associated with the isospin density fluctuation. In the case of ANM, because natural symmetries are lost, these relations break down where one can show that [7]

$$\frac{\partial P}{\partial\rho_1}|_{T,y} = \frac{\lambda^+}{\sqrt{t}}(t \cos \beta + \sin \beta)^2 + \frac{\lambda^-}{\sqrt{t}}(t \sin \beta - \cos \beta)^2 \quad (6)$$

$$\frac{\partial\mu_p}{\partial y}|_{T,P} = \rho_n \lambda^+ \lambda^- \left( \frac{\partial P}{\partial\rho_1}|_{T,y} \right)^{-1} \quad (7)$$

where  $\beta = 1/2 \tan \mathcal{F}_{np} / (\mathcal{F}_{pp} - \mathcal{F}_{nn})$  and  $t = \rho_n N_0^n / \rho_p N_0^p$ . These two equalities illustrate explicitly that the simplicity of SNM is not preserved in ANM. Hence, one should come back to eigen analysis of the curvature matrix  $[\mathcal{F}_{ij}]$ . We propose a generalisation of the definition of isoscalar and isovector modes in ANM. According to Fig. 5, the instability is of isoscalar type if its direction points in the direction of the first bissectrix with an absolute value angle less than 45 degrees, while it is of isovector kind if its direction points in the direction of the second bissectrix with an absolute value angle less than 45 degrees.

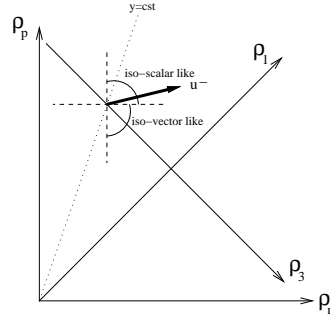


Figure 5. Here we illustrate the generalisation of the definition of isoscalar and isovector modes.

## 5. Results

The local stability condition in ANM is expressed by Eq. 4. If it is violated the system is in the unstable region of a phase transition. Two cases are then possible : i) only one eigenvalue is negative and one order parameter is sufficient to describe the transition or ii) both eigenvalues are negative and two independent order parameters should be considered meaning that more than two phases can coexist.

For ANM below saturation density, the case ii) never occurs since the asymmetry energy has always positive curvature ( $\mathcal{F}_{33}$ ). Indeed, the asymmetry term in the mass formula behaves like  $(N - Z)^2$  times a positive function of  $A$  showing that the dominant  $\rho_3$  dependence of the asymmetry potential energy is essentially quadratic and that  $\mathcal{F}_{33}$  is a positive function of the total density. Recent Bruckner calculations in ANM [9] have confirmed the positivity of  $\mathcal{F}_{33}$ . They have parameterized the potential energy with the simple form  $\mathcal{V}(\rho_1, \rho_3) = \mathcal{V}_0(\rho_1)\rho_1^2 + \mathcal{V}_1(\rho_1)\rho_3^2$  with  $\mathcal{V}_0/\mathcal{V}_1 \sim -3$  and  $\mathcal{V}_0 < 0$ . This is also true for effective forces such as Skyrme forces. For example, the simplest interaction with a constant attraction,  $t_0$ , and a repulsive part,  $t_3\rho_1$ , leads to  $\mathcal{V}(\rho_1, \rho_3) = (3\rho_1^2 - \rho_3^2)\beta(\rho_1)$  with  $\beta(\rho_1) = (t_0 + t_3/6\rho_1)/8$ . The function  $\beta(\rho_1)$  is negative below saturation density, hence the contribution of the interaction to  $\mathcal{F}_{33}$  in the low density region is always positive.

These arguments show that, below saturation density, the  $\rho_3$  curvature,  $\mathcal{F}_{33}$ , is expected to be positive for all asymmetries. Since the curvature in any direction,  $\mathcal{F}_{ii}$ ,

should be between the two eigenvalues  $\lambda^- \leq \mathcal{F}_{ii} \leq \lambda^+$  we immediately see that if  $\mathcal{F}_{33}$  is positive one eigen curvature at least should remain positive. In fact for all models we have studied  $\mathcal{F}_{33}$  appears to be always large enough so that the trace is always positive demonstrating that  $\lambda^+ > 0$ . Since  $\text{Tr}[\mathcal{F}_{ij}] = \mathcal{F}_{nn} + \mathcal{F}_{pp}$ , this can be related to the positivity of the Landau parameter  $\mathcal{F}_{nn}$  and  $\mathcal{F}_{pp}$ .

The large positive value of  $\mathcal{F}_{33}$  also indicates that the instability should remain far from the  $\rho_3$  direction i.e. it should involve total density variation and indeed we will see that in all models and for all asymmetry the instability direction hardly deviates from a constant asymmetry direction ( $\delta\rho_3 \ll \delta\rho_1$ ). This isoscalar nature of the instability can be understood by looking at the expression of the eigen-modes in the  $(\rho_n; \rho_p)$  coordinates. Since  $\lambda^- \leq \mathcal{F}_{ii} \leq \lambda^+$ , the differences  $\lambda^- - \mathcal{F}_{ii}$  is always negative demonstrating, using Eq. 3, that the instability is of isoscalar type, if the Landau parameter  $\mathcal{F}_{np}$  is negative. Hence, there is a close link between the isoscalar nature of the instability and the attraction of the proton-neutron interaction [7].

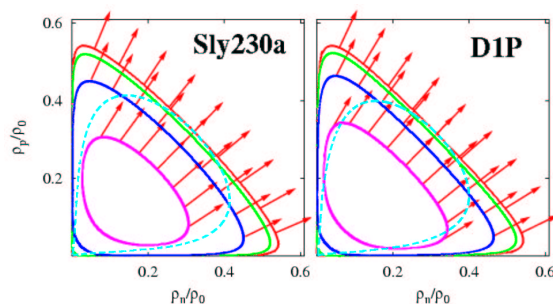


Figure 6. This is the projection of the iso-eigen values on the density plane for Sly230a (left) and D1P (right). The arrows indicate the direction of instability. The mechanical instability is also indicated (dotted line).

Various contour of equal imaginary sound velocity are represented in Fig. 6 for SLy230b and D1P interactions. The more internal curve correspond to the sound velocity  $i0.09c$ , after comes  $i0.06c$ ,  $i0.03c$  and finally 0, the spinodal boarder. For these two recent forces which takes into account pure neutron matter constraints, the predicted instability domains are rather similar. We observe that in almost all the spinodal region the sound velocity is larger than  $0.06c$ .

Let us now focus on the direction of the instability. If  $\delta\rho^-$  is along  $y=\text{cst}$  then the instability does not change the proton fraction. For symmetry reasons pure isoscalar ( $\delta\rho_3 = 0$ ) and isovector ( $\delta\rho_1 = 0$ ) modes appears only for SNM so it is interesting to introduce a generalization of isoscalar-like and isovector-like modes by considering if the protons and neutrons move in phase ( $\delta\rho_n^- \delta\rho_p^- > 0$ ) or out of phase ( $\delta\rho_n^- \delta\rho_p^- < 0$ ). Fig. 6 shows the direction of instabilities along the spinodal boarder and some iso-instability lines. We observed that instability is always almost along the  $\rho_1$  axis meaning that it is dominated by total density fluctuations even for large asymmetries. Fig. 7 presents



the angle of the eigen state  $\delta\rho^-$  with the isoscalar axis normalized by the angle between the  $y=\text{cst}$  line and the isoscalar axis (denoted  $\chi$ ) for 3 models (D1P, SGII, SLy230a). We can see that this quantity is in between 0 and 1, so that the instability direction is between the  $y=\text{cst}$  line and the  $\rho_1$  direction. This shows that the unstable direction is of isoscalar nature as expected from the attractive interaction between proton-neutron. The total density is therefore the dominant contribution to the order parameter showing that the transition is between two phases having different densities (i.e. liquid-gas phase transition). The angle with the  $\rho_1$  axis is almost constant along a constant  $y$  line. This means that as the matter enters in the spinodal zone and then dives into it, there are no dramatic change in the instability direction which remains essentially a density fluctuation. Moreover, the unstable eigenvector drives the dense phase (i.e. the liquid) towards a more symmetric point in the density plane. By particle conservation, the gas phase will be more asymmetric leading to the fractionation phenomenon. Those results are in agreement with recent calculation for ANM [7] and nuclei [19].

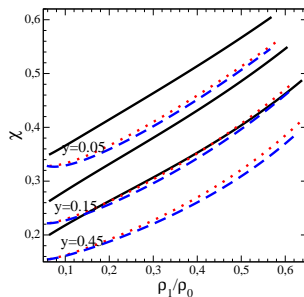


Figure 7. Angle of  $\delta\rho^-$  with the isoscalar axis normalized by the angle between the  $y=\text{cst}$  line and the isoscalar axis (denoted  $\chi$ ) for various values of  $y$  noted on the figure and for 3 models (solid: SLy230a, dotted: D1P, dashed: SGII).

We want to stress that those qualitative conclusions are very robust and have been reached for all the Skyrme and Gogny forces we have tested (SGII, SkM\*, RATP, D1, D1S, D1P,...) including the most recent one (SLy230a, D1P) as well as the original one (like SIII, D1).

## 6. Conclusion

In this paper, we have shown that ANM does not present two types of spinodal instabilities, a mechanical and chemical, but only one which is dominantly of isoscalar nature as a consequence of the negativity of the Landau parameter  $\mathcal{F}_{np}$ . This general property can be linked to the positivity of the symmetry energy curvature  $\mathcal{F}_{33}$ . This means that the instability is always dominated by density fluctuations and so can be interpreted as a liquid-gas separation. The instabilities tend to restore the isospin symmetry for the dense phase (liquid) leading to the fractionation of ANM. We have

shown that changing the asymmetry up to  $\rho_p < 3\rho_n$  does not change quantitatively the density at which instability appears, neither the imaginary sound velocity compared to those obtained in SNM. All the above results are not qualitatively modified by the temperature which mainly introduce a reduction of the spinodal region up to the SNM critical point where it vanishes. The quantitative predictions concerning the shape of the spinodal zone as well as the instability times depends upon the chosen interaction but converge for the various forces already constrained to reproduce the pure neutron matter calculation.

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